

# Radionuclide Capture Using Membranes

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## Abstract<sup>1</sup>

Much of the radioactivity in nuclear wastes is in the form of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . These elements can be separated into a much smaller volume by ion exchange, reducing the volume of wastes that must be stored. New technology has been developed to incorporate almost any sorbent particle into membranes and then fabricate these into chemical absorbing cartridges. These cartridges can be used to capture radionuclides in a form that is easy to handle and either process further or store. Advantages over columns include more complete decontamination, much higher throughput, and final waste forms which take up less volume and can be handled with less worker exposure to radiation.

In early 1989 3M introduced this technology as a form of solid phase extraction (SPE), which is used to remove dissolved species from water. The sorbent is loaded into a web or membrane which is used in a filtration-like process. Several classes of materials have been incorporated into 3M membranes, including organic ion exchange materials, inorganic adsorbents, zeolite structures and macrocyclic molecular recognition compounds. The resulting membranes are characterized by high separation efficiencies, high radionuclide loading, fast flow rates and kinetics and physical ruggedness. A radiolytically resistant material may be used as the membrane matrix. Because the membrane is very densely packed with small (5-25 $\mu\text{m}$ ), high surface area particles the flow rate can be 10 to 100 times faster than standard column ion exchange processes while achieving equal extraction efficiencies. Particles of this size would result in unacceptable back pressure if used in columns. This membrane separation technology allows the use of a number of known, high performance, chemical adsorbing powders, which previously could not be put into a useful engineered form because of their small particle size.

Membranes and cartridges for radionuclide capture have been tested at Idaho National Engineering Laboratory (INEL), West Valley Nuclear Services(WVNS), and Hanford. Demonstrations are planned at the Savannah River Site(SRTC) and Argonne National Laboratory

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(ANL). Results to date are summarized below.

Laboratory scale experiments at Idaho National Engineering Laboratory (INEL) showed that strontium and cesium could be removed from actual radioactive acidic waste even in the presence of relatively high concentrations of other ionic species. The membranes used were 25 mm diameter, 0.5 mm thick filter-type disks with either macrocyclic molecular recognition particles for the removal of strontium and lead, or crystalline titanium phosphate (PhTiA) particles for the removal of cesium.

A larger scale demonstration was conducted at INEL, Test Area North (TAN), where a groundwater treatment facility (GWTF) had been built to treat injection well water contaminated with both organic solvents and radionuclides. 1,250 gallons of water were processed at a flow rate of 0.25 gallons per minute through a test train containing filters and cartridges of sodium titanate for strontium and potassium cobalt hexacyanoferrate (KCOHEX) for cesium. Relatively high concentrations of calcium and magnesium caused 50% breakthrough ( $[Sr]_{out}/[Sr]_{in} = 0.5$ ) after approximately 800 gallons of water had passed through the Sr absorber. Cesium did not breakthrough for the entire demonstration, with a detection limit of 9 pCi/L.

Trace quantities of technetium (as an anionic species) and cesium were removed from process water at the West Valley Nuclear Services site in New York state. More than 1,500 gallons of water from a storage lagoon were processed continuously at a flow rate of about 0.1 gallon per minute. The original technetium level was reduced by more than an order of magnitude, with 50% breakthrough ( $[TcO_4]_{out}/[TcO_4]_{in} = 0.5$ ) being observed after about 90 gallons. A decontamination factor (DF) of at least 26 was achieved for cesium removal, reducing the level from  $1.17 \times 10^{-7}$  FCi/ml to less than the detection limit,  $4.3 \times 10^{-9}$  FCi/ml. Sorbent used for the removal of technetium was a carbon membrane cartridge loaded with Aliquat<sup>TM</sup> 336, an anion capturing material. A cartridge loaded with KCOHEX was used to remove cesium. An effective prefiltration system was used to remove algae.

In subsequent work, a proprietary absorber was used to remove pertechnetate. A slip stream was processed at 0.3 gallons per minute, treating more than 3,200 gallons over 192 hours. The initial technetium concentration was  $8.6 \times 10^{-7}$  FCi/ml. A DF of 100 was achieved at the beginning of the run and leveled off at about ten in the last several hours. Cesium was not detected in the effluent of the test system at any time during the experiment. Initial cesium level was  $3.8 \times 10^{-7}$  FCi/ml, meaning that 4.7 FCi of  $^{137}\text{Cs}$  were collected by the cartridge.

The prototype of equipment scalable to run at 50 gallons per minute was evaluated at "N" Springs and "N" Basin, Hanford for the removal of strontium and cesium. Operating at flow rates ranging from 1.0 to 3.5 gallons per minute the equipment performed as designed. Separations of strontium and cesium which were achieved were those predicted based on the results of tests at INEL and in the laboratory at 3M using samples of the challenge water.